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Ionic Liquid Mixed Polymer Electrolyte for Supercapacitor

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In present paper the authors targeted to present the recent Ionic Liquids doped polymer electrolyte and tested its electrochemical, thermal, and physicochemical properties, which are decisive for Electrochemical devices. Ionic Liquids can also be considered designer solvents to substitute the more flammable organic carbonates and enhance the green credentials and performance of renewable energy storage devices mainly Supercapacitor and Lithium Ioin Battery. This review presents the recent Ionic Liquids doped electrolyte electrochemical, thermal, and physicochemical properties, which are decisive for supercapacitors and lithium-ion batteries. Ionic Liquids can also be considered designer solvents to substitute the more flammable organic carbonates and enhance the green credentials and performance of renewable energy storage devices like Supercapacitor, Batteries.

1. Introduction

In literature different materials have been reported to resolve the energy problem that the world is facing in everyday life. Supercapacitors have attracted more than batteries due to their interesting properties such as fast charge (1-10s) and better cycle stability. Generally, solid materials are categorized into three principal groups: metals, ceramics, and polymers. Therefore, they are used to fabricate electrochemical storage and conversion devices. A great demand great in materials has been noticed in designing electrochemical energy storage devices.[1,2] Mainly, supercapacitors (SCs) attracted more than batteries due to their interesting properties such as fast charge (1-10s) and better cycle stability.[3] To achieve better capacitance than conventional supercapacitors, SCs use electrode materials with high surface area and thin electrolytic dielectrics.[4–8] This will increase energy densities since the power density of conventional supercapacitors remains unchanged. Consequently, SCs can be combined with another storage device to improve power efficiency and cycle life or used alone in many technological applications and automobiles.[9–11]

During the 1957–1990s, SC technology was low in demand compared to hybrid electric vehicles. Now, this technology targets enhancing electrode materials' performance to make SCs more competitive than batteries with high energy density.

Now, this technology targets enhancing electrode materials' performance.[12–14] Various types of SCs are used in the drive systems to fully supply their braking systems with power and energy.[15,16]

Therefore, SCs are reliable in many storage devices because of their ability to protect them against power disruptions.[13,17,18] Their performance relies significantly on the electrode-electrolyte and the potential window between electrodes.[19] Researchers are interested in developing efficient materials for electrodes in SC with an appropriate structure to ease ion diffusion.[20] Deep learning of storing energy in SCs requires a good choice of electrode materials and electrolytes to make them more competitive than batteries with high energy density.

1.1. Supercapacitors as Emerging Energy Stores

SCs have many characteristics such as high power density, good stability, and cycle life, which make them useful in many electronic-powered devices and power devices.[21] Their main drawback is energy storage capacity which limits their use on a large scale. Without including some important parameters, the variation of power density with energy is shown in **Figure 1**. SCs show a much higher cycle life and exceptionally low recharge times compared to batteries.[22]

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DOI: 10.1002/masy.202400153

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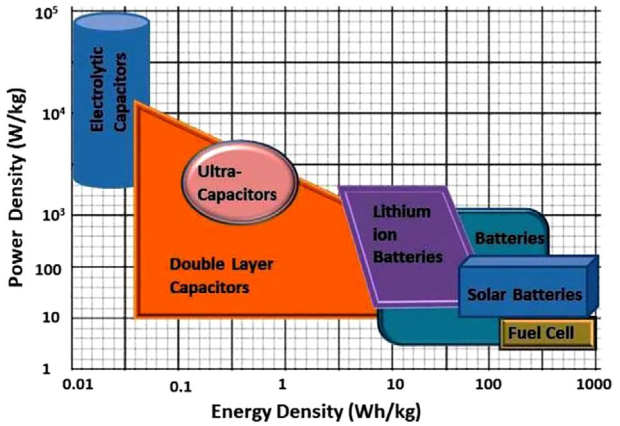

Figure 1. Ragone plot showing power versus energy density for different energy storage devices.[23]

The performances of conventional capacitors, supercapacitors, and batteries are evaluated by key parameters such as cycle life, specific energy charge/discharge time, and storage mechanism. Thermal stability is one point into rest that reflects good storage devices, and capacitance is one of the most crucial parameters to get the energy density evaluated. This parameter relies on the chosen materials and electrolytes used during the electrochemical process. SCs compared to batteries, can be stable in the range [−40, 100 °C],[24] compared to batteries which can be only functioned between −20 and 60 °C. Capacitance is one of the most crucial parameters to get the energy density evaluated. To ameliorate the performance of the device, scientists have developed many methods to enhance the capacitance such as the fabrication of high surface area nanoporous materials[25,26] like coating Graphite oxide with MnO_2 nanorods homogeneously. This technique is effective not only in reducing the ionic diffusion path in the charge /discharge process but also in promoting a good charge transfer; (b) mass transportation is highly improved using active materials; (c) the increase in wettability of the electrode surface with electrolyte (d) setting the mechanisms of mass transport.[27] These ways lead to shortening ESR, ameliorating the penetration of electrolyte ions into the active material, and then enhancing the specific capacitance, charging process, and electrode materials.

1.2. Categories of Supercapacitors

Supercapacitors are generally divided into three groups: EDLCs, pseudo capacitors, and hybrid capacitors. **Figure 2** shows the different types of supercapacitors with their electrode materials. The main of this study is to present the different materials and components used in SCs and their effect on the device performance.

2. Materials and Components for Supercapacitors

2.1. Energy Storage Mechanisms in Supercapacitors

2.1.1. Electrical Double Layer Capacitors (EDLCs)

EDLCs are supercapacitors that store energy from the interface between the electrode material and electrolyte. They have a higher energy density than conventional capacitors because of their small space between the charged plates. EDLCs Compared to conventional capacitors contain an electrolyte (such as H_2SO_4, KOH, or Na_2CO_3) that creates dielectric between the two plates as illustrated in **Figure 3**. Conventional capacitors suffer from unsuitable space between the two charged plates and their geometry which limits their performances.

In the 19th century, Helmholtz designed the first EDLC concept based on colloidal particle interfaces. Gouy and Chapman changed it into the diffusive layer model and Stern proposed a new concept. (**Figure 4A**) based on the colloidal particle interfaces and was changed into the diffusive layer model by Gouy and Chapman (Figure 4B). Generally, the capacitive behavior in EDLCs is the same with parallel plate capacitors. Therefore, the capacitance rises when point charges are close enough to the electrode surface. A new concept which combines both an inner region also named by stern layer and a diffusive layer, was proposed by Stern (Figure 4C).

In this model, the solid stern layer contains such anions and nonspecific adsorbate counter-ions that are not specific. The EDLC capacitance (C_{dl}) is obtained by combining the capacitance in the Helmholtz model (C_H) and differential capacitance (C_{diff}) for both the Stern layer and diffusive region respectively as given below:

$$\frac{1}{C_{dl}} = \frac{1}{C_H} + \frac{1}{C_{diff}} \quad (1)$$

In that model, the overall capacitance in EDLC, C_{dl} was considered as a series of capacitance with C_H and C_{diff}. In EDLCs, the electrical conduction mechanism at the level of the electrode surface is influenced by chemical affinities in the electric behavior. EDLCs are strongly conditioned by the porosity of electrodes, which deliver a high specific area. EDLCs fabricated with porous carbon conducting electrode material can be represented as shown in **Figure 5**.

The EDLC-specific capacitance is calculated using the Equation 2 and mechanical stability.[31] The Faradaic reaction which reflects the mechanism of charge transfer takes place when the electrolyte and the solid materials interact together. CP material-based electrodes contribute to improving the electric current and exhibit maximum electronic conductivity.

$$C = \frac{\varepsilon_r \varepsilon_0 S}{l} \quad (2)$$

where ε_r is the dielectric constant, ε_0 is absolute permittivity, S *is* the specific area of electrodes for ions absorption, and l is the effective thickness of the EDLC.

This relationship reflects the unapproachable association between micropores and ion sizes on the surface as reported by Raymundo-Pinero and co-workers.[32] The relationship between capacitance and pore size has been investigated in many studies.[33–36] It is believed that a pore size of 1 nm is small enough for hydrated ions.[37]

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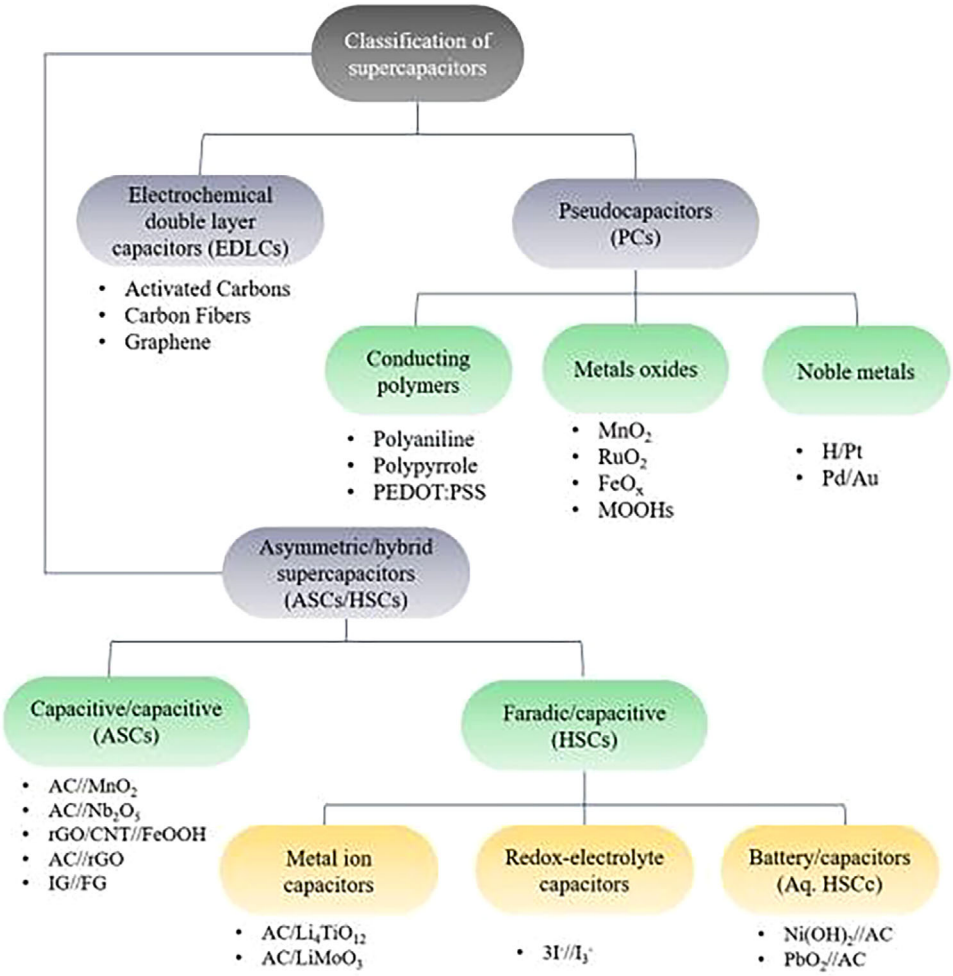

Figure 2. Classification of supercapacitors with their representative electrode materials based on the energy storage mechanism that occurs in each type of SC.[28]

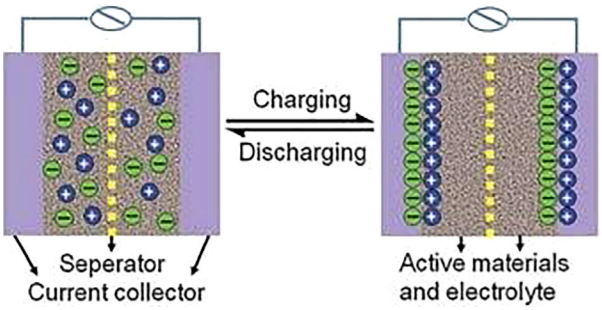

Figure 3. The charge storage mechanism of EDLC.[29]

2.1.2. Pseudo-Capacitors

Pseudo-capacitors (PCs) rely on the Faraday charge process, which takes place when the electrolyte and the solid materials interact together.

This process can be reversible irreversibly, the chemical reactions (oxidation/ reduction) do not generate any new chemical species in the reversible process whereas in, the irreversible process, new species are produced. PCs depend significantly on particle sizes and morphologies.[38,39] An extrinsic pseudocapacitor consists of nanostructured surfaces. During the ion storage process in the bulk material, the phase transformation prevents the presence of Extrinsic PC.[40] Therefore, PC materials are distinguished from battery-type materials using electroanalytical experimental materials such as carbons, conducting polymers (CPs), and metal oxides[41,42] are widely employed pseudocapacitors, the capacitance can be higher than EDLC of 10 to 100 times but unfortunately with a powerful performance lower than that of an EDLC because of shrinking and swelling exhibit during the charge-discharge mechanism, that reduce the cycle life.

2.1.3. Hybrid Capacitors

SCs are a middle ground between batteries and capacitors. The combination of EDLC and PCs forms Hybrid capacitors, and there is no big difference between them (EDLC and PCs) only the storage principle. There is no chemical reaction on the electrodes in the EDLC device. Otherwise, the oxidation/reduction reactions take place just on the electrode surface of PCs which shows

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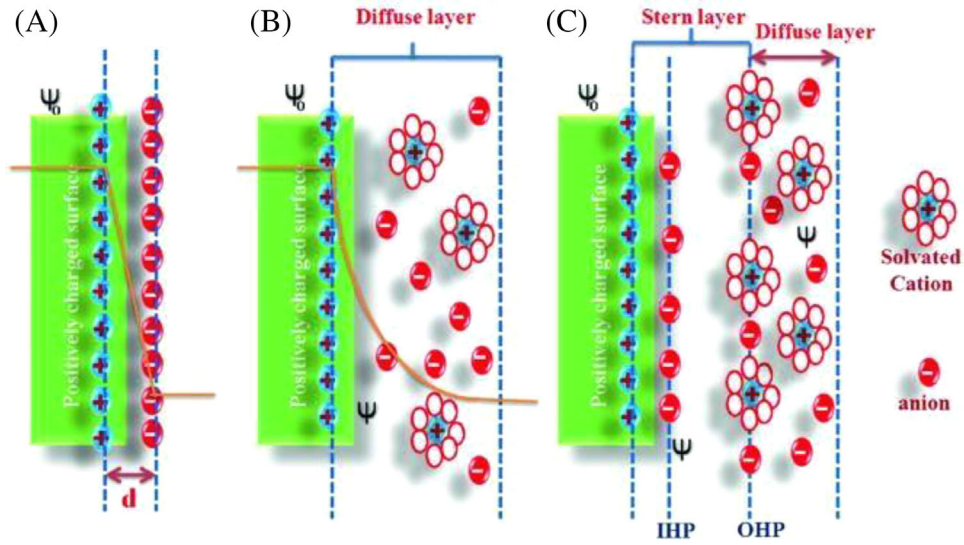

Figure 4. Different models were developed by stern layer at the positively charged surface: (A) the Helmholtz model, (B) the Gouy–Chapman model, and (C) the Stern model.[30]

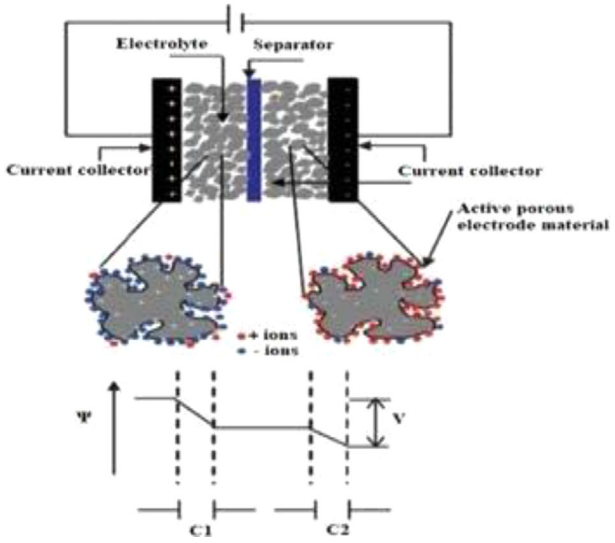

Figure 5. Porous electrode material-based EDLC.[30]

higher capacitance than EDLCs. The enhancement of EDLC capacitance may be done by making nanosized electrode material with high porosity and an ameliorated energy density. Asymmetric hybrid SCs can be fabricated utilizing Pseudo Capacitor and EDLC materials into the positive and negative electrodes respectively. These SCs have been reported to show better specific capacitances than carbon electrodes. We distinguish two categories of asymmetric/hybrid SCs: the first one consists of both capacitor-type electrode and capacitor-type electrode SCs and the second type is based on capacitor-type electrodes and battery-type electrode SCs.[43,44] Cong et al.[45] claimed SCs based on PVA-H_2SO_4 gel as an electrolyte with electrodes have been reported. They revealed the graphene-based SCs sandwiched with PANI show a high capacitance of ~763 Fg^{-1} at 1 Ag^{-1} in 1M H_2SO_4 compared to those based on a single electrode component. Recently, it has been proved that utilizing TCMs such as nitrides, metal oxides sulfides and carbides can provide better specific capacitances than carbon electrodes. This was also supported by the results of Binitha et al.[46] which revealed distinctive capacitance of 256 F/g and 102 F/g using fabricated porous α-Fe_2O_3 fiber and nano grain structures respectively, with 1 M LiOH as an electrolyte in SCs.

Li-ion batteries take considerable time to charge compared to SCs which only take a few times(s). The association of the fast charge of SCs and Li-ion batteries with high energy is conducted in the manufacturing of hybrid capacitors.[47,48] Hybrid capacitors based on carbon nanofillers/boron-doped diamond electrodes have been manufactured using 1 M H_2SO_4 as electrolyte and 1M Na_2SO_4 aqueous solution (0.05M Fe(CN)6 3-/4-) as reducing species.[49] Many solutions are proposed to increase the surface area (a) amelioration of the electrode conduction;[50] (b) Use the nano-architectures to reduce the track of diffusion on electrodes;[51] (c) reduce speeding the diffusion of ions into all directions;[52] Therefore, various materials such carbon-based materials with sp2 and sp3 atomic orbital hybridizations have been widely tested to meet a great demand such as fullerenes, graphite, graphene, nanotubes, diamond, and so on with their dimensions (0D–3D).[53]

EDLCs use inert electrolytes, whereas Pseudo capacitors use electrolytes based on redox species. One approach to get efficient SCs is by adding soluble redox species to the electrolyte. Therefore, the charge storage capacity depends strongly on the number of redox species but is not enough to completely understand the performances of SCs.

2.2. Electrodes Material Used for Supercapacitors

Materials that are used in SC electrodes define the performance and the storage mechanism in SCs as discussed previously. **Table 1** below the different materials used for supercapacitors.

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Table 1. Classification of materials for supercapacitor electrolytes.

Type of Devices	Electrode Materials
	Activated carbons (AC)
EDLCs	Carbon nanotubes (CNT)
	Carbon fibers (CF)
	Carbon aerogels (CAs)
PCs	Conducting polymers (CPs)
	Metal oxides (MO)
Hybrid supercapacitors	
Asymmetric	Carbon materials (CMs), metal oxides (MOS)
Composite	Carbon materials, conducting polymers
Battery-type	

Carbon is the most useful material for supercapacitors, and it can be transformed in many forms.

The SC capacitance changes with the effective surface area of the materials. However, the effective area is not always responsible for the interaction on the electrode surfaces. This involves the capacitance of selected electrode material-based EDLC can also change nonlinearly with the effective surface area.[54]

Largeot and co-workers suggested that the ions size and pores size both should be close enough to higher the capacitance in the double-layer capacitor.[55] Otherwise, the shortest pores contribute to reducing the capacitance. The capacitance dependence on pore size can be shown in **Figure 6**.

It is obvious to note that the capacitance of a material is affected by its porosity; In fact, the capacitance is reduced as the size of the pore increases due to the increase in the distance between the center of the associated ions and the pore walls. The capacitance of a material is affected by its porosity, and the use of several active materials in EDLCs and PC devices showed their impact on the specific capacitance.[56]

2.2.1. Carbon Materials

(i) Activated Carbons (ACs)

ACs are materials commonly used in EDLC supercapacitors and are well known for their highly porous structure. Although highly porous materials are better for obtaining a good specific capacitance, pores with sizes smaller than ion sizes do not contribute to the charge storage as reported by Shi (1996), and Qu and Shi (1998). In the same order, Frackowiak and Beguin (2001) revealed that high porosity can reduce the conductivity limit the maximum power density, and create a distributed electrolyte resistance that restrains charge/discharge rates. In addition, this may create a distributed electrolyte resistance that restrains charge/discharge rates as reported by Pandolfo and Hollenkamp (2006). Considerably advanced techniques were found to improve the surface area of ACs up to 3000 m^2 g^{-1} with their performance.[57] The electrical properties of EDLC vary accordingly with the EDLC performance which relies on the pore size distribution, surface area of electrode material, structural morphology, and conductivity as reported by Ba et al.[58]

(ii) Carbon Nanotubes (CNTs)

CNTs are highly applicable in supercapacitors as electrode materials because of their good chemical, optical, mechanical, and electronic features. CNTs have a large surface area, good electrical conductivity, are lightweight, have good flexibility, and exhibit high polarizability. On the production market, CNTs are inexpensive, available, and durable. The continuous charge distribution in CNT-based supercapacitor electrodes with a large surface area achieves good capacitances compared to that of those based on AC electrodes as reported by Frackwiak and Beguin 2001 and Frackwiak et al. 2000. CNTs are mostly used because of their good electrochemical properties such as good electrical conductivity (10^{-5} S cm^{-1}), lightweight, good flexibility, and which exhibit high surface area (1600 m2g^{-1}).[59,60] In addition, they possess electrodes with high polarizability and several investigations

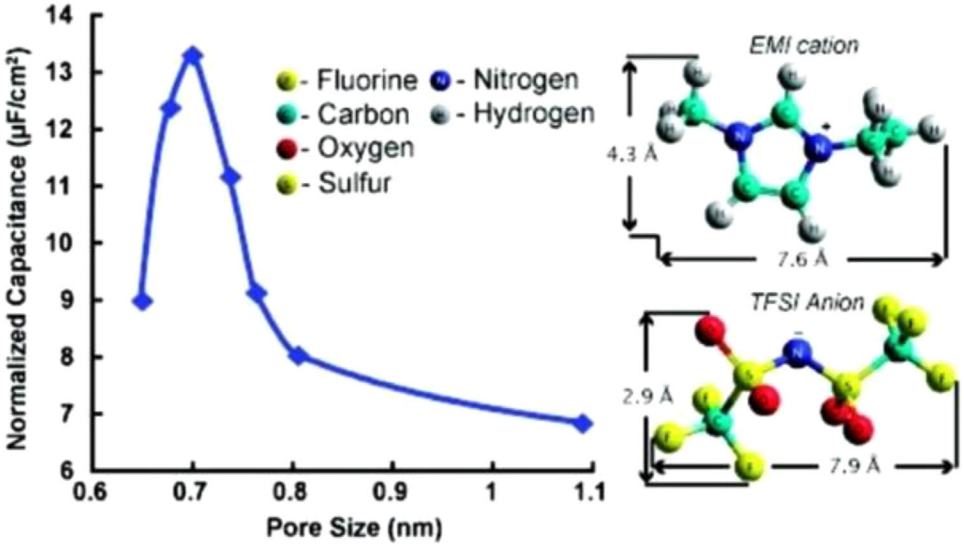

Figure 6. Variation of normalized capacitance with pore size of.[55]

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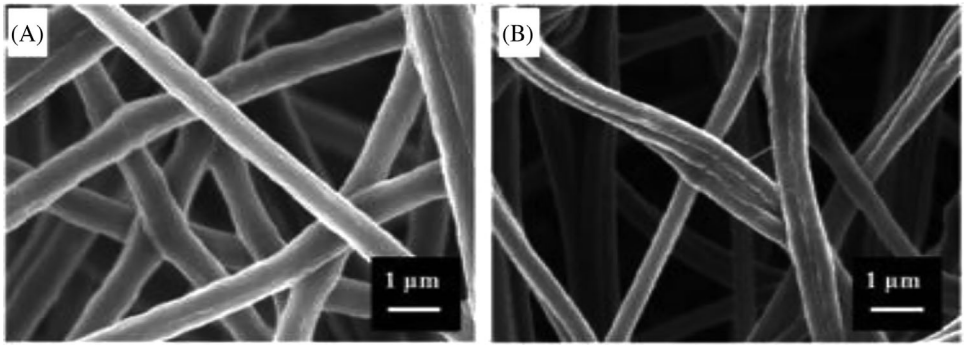

Figure 7. Structural morphology of (A) polyaniline and (B) polyaniline doped carbon nanotube.[69]

have shown the use of single-walled CNT (SWCNT) as electrode material for SC application.[61]

(iii) Carbon Aerogels

These materials are based on interspaced mesoporous, derived from carbon nanoparticles, and are especially used in EDLC supercapacitor electrodes without a binder material. Carbon aerogels-based electrodes contribute to increasing the power performance due to the lower equivalent series resistance, as approved by Arico et al. (2005).

2.2.2. Metal Oxides

(i) Ruthenium Oxide (RuO_2)

Metal oxides (MOs) exhibit high conductivity and are widely used in supercapacitor electrodes because of their long operation time and large specific capacitance. In 2007, Lee et al. revealed that RuO_2 has a considerable interest in supercapacitor technology. Zhen and Jow (1995) reported that the hydrous form of RuO_2 helps to exhibit high capacitance better than both carbon materials and CPs. However, the high cost of RuO_2 limits its production and availability on the market. Additionally, RuO_2 combined with other materials in SCs has been extensively investigated.[62] However, RuO_2-based energy devices deliver a high specific capacitance of 905 F g^{-1} but the high cost limits its production and availability on the market.[63]

(ii) Manganese Oxide (MnO_2)

MnO_2 is a good candidate for supercapacitor electrodes, but it has poor conductivity. Several techniques have been employed to synthesize these compounds, and they have been employed in composite form with CNTs to make them more useful. Therefore, Wang et al. in 2005; Xie and Gao in 2007 reported in their works this material has been employed in composite form with CNTs to make it more useful. Several investigations have been done on the mesoporous nature of various oxides to be useful as a material for SC electrodes.[64]

2.2.3. Conducting Polymers

Organic polymers which conduct electric current through a conjugated bond matrix are called conducting polymers (CPs). CPs deliver such energy density better than metal oxides, with good reversible capacity and low cost.[65] Several CPs such as PPy, PANI, and PTs have been investigated which show a high conductivity.[66] In previous works, CPs were studied in many forms such as nanowalls, nanosheets, and nanorods, and used in SCs to optimize their performance.[67] CPs based on nano architectures present many advantages such as high surface area and high porosity, directly leading to improved SC performances. Recently, one-dimensional nano-structures-based CP materials were utilized to achieve better performance in PCs than their bulk counterparts.[68] **Figure 7** shows the morphological structure of polyaniline and polyaniline-doped Carbon nanotubes used as conducting polymers in supercapacitors.

2.2.4. Composites

Carbon materials such as graphene and CNTs are intensively utilized in electrodes even showing a low specific capacitance.[70] In contrast, electrode materials such as CPs and metal oxides produce efficient PCs that can deliver high energy.[71] Composite materials are most of the time employed in hybrid SCs as electrode materials with high energy and power densities. They involve the combination of carbon materials with either CPs or MOs. Farack and co-workers (2006) and Jurewicz and co-workers (2001) reported that supercapacitor electrodes fabricated from CNT and Polpyrrole combinations exhibit higher capacitance to electrodes than those entirely made of polypyrroles or CNTs. All these electrode materials are synthesized using several techniques, but each method presents its advantages and drawbacks.

2.3. Electrolytes

The electrolyte is a key element of electrochemical supercapacitors (ESs), which results in the compensation of charges on both electrodes in a device and gives ionic conductivity.[72] The electrolyte is a key element of electrochemical supercapacitors, and it plays a crucial role in the creation of the double layer and PCs.

The choice of an electrolyte depends on the electrode materials, ionic size and type, ion-solvent interaction, concentration, and electrochemical stability potential window.

Therefore, electrolytes can modify the cycle life, the capacitance, the pseudo capacitance of an EDL, and power density.

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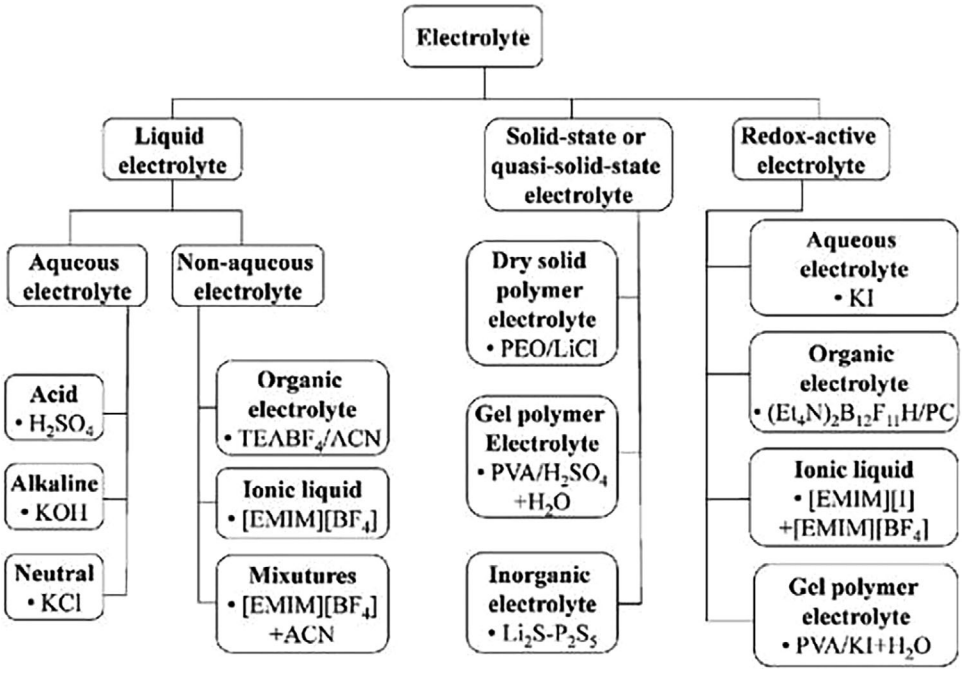

Figure 8. The types of electrolytes used in electrochemical SCs. Aqueous, organic, and ionic liquid electrolytes have advantages and disadvantages, and new electrolyte materials have been developed to optimize the device's performance.[72]

As an example, the operating voltage is measured by the ESPW which directly influences the cell performance.[72]

In the literature, many electrolytes are used for energy storage devices. They are principally categorized into two states: liquid and solid /quasi-solid-state electrolytes.[73] The illustration is given below in **Figure 8**.

Therefore, liquid electrolytes are subdivided into aqueous, organic, and ionic liquid electrolytes whereas solid or fluid electrolytes are available in organic and inorganic forms. These two groups of electrolytes have some advantages and disadvantages. Aqueous electrolytes can offer high conductivity and capacitance but show a narrow working voltage. Ionic liquids and organic electrolytes can operate at elevated voltages, but they show a lower conductivity. Solid-state electrolytes elude the problem of leakage, which is considered a mutual problem for liquid electrolytes but suffers from low conductivity compared to liquid-state electrolytes. New electrolyte materials have been developed to optimize the device performance with (a) higher ionic conductivity, (b) a wider stability potential window, (c) a large range of operating temperature, and (d) good thermal stability.[74] However, the investigation of the properties of these electrolytes using the experimental advanced approach aids in better comprehending the crucial influence of electrolytes on the SC properties.[75]

2.3.1. Aqueous Electrolytes

In general, aqueous electrolytes exhibit better conductivity than that of organic electrolytes and ILs. This leads to enhanced power delivery of ESs with a decrease in ESR.

Aqueous electrolytes have a low electrochemical stability potential window, so most commercial ESs are based on organic electrolytes instead of aqueous ones. However, aqueous solutions are showing a main drawback of low electrochemical stability potential window, for this reason, most of the common commercial ESs are based on organic electrolytes instead of aqueous ones. Usually, the common aqueous electrolytes such as H2SO4, Na2SO4, and KOH are commonly formed from acid, alkaline, and neutral solutions.[75] MnO_2 nanorods were used as electrode materials and neutral aqueous solutions were used as electrolytes in a newly prepared AC supercapacitor. They exhibit good cycling behavior, a high energy of 17 Wh kg^{-1}, and a high power density of 2 kW kg^{-1}.[76] Scientists are using electrolytes in SCs because of properties such as high capacitance, ease of manipulation, and no leakage. Researchers experienced slightly modifying aqueous electrolytes by adding various gels to stabilize the electrolytic system.[77] In previous works, solid-state electrolytes have become promising candidates for energy storage device applications. The novel solid polymer electrolyte of PVA and GO showed excellent performance after a month's test. Pan and coworkers prepared a novel solid polymer electrolyte of PVA and GO;[78] after filtering the system with an aqueous electrolyte the system reached excellent performance. This system showed less decay of ionic liquid (38.4%) compared to 84% for PVA after one month of testing in a relative humidity atmosphere of 45%. This demonstrated that aqueous electrolyte-based electronic apparatus can offer good stability of electrical performance.

2.3.2. Organic Electrolytes

Organic electrolytes are currently used in ESs and EDLCs because they possess a high potential window, but they are volatile, toxic, flammable, and costly.[72] In EDLCs, the organic

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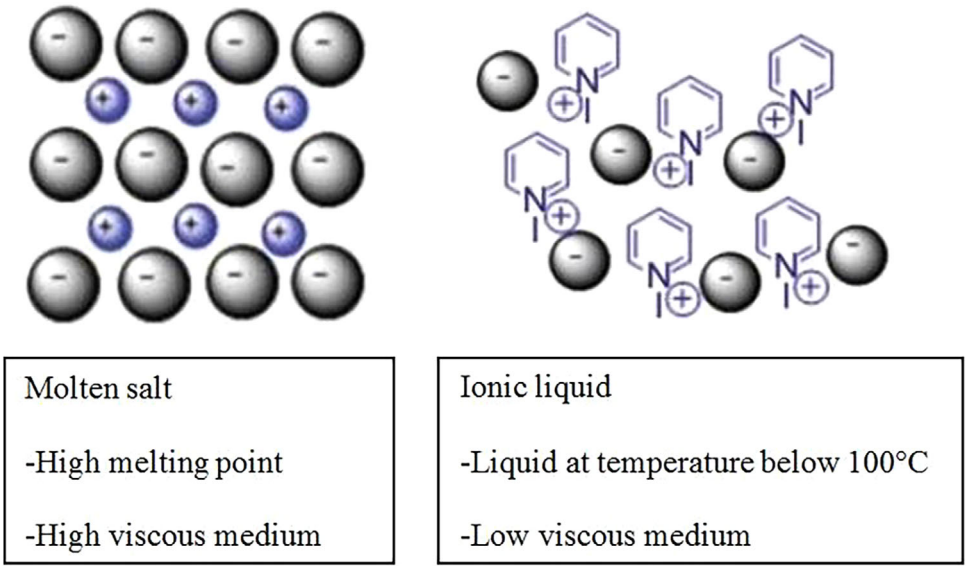

Figure 9. Structure of molten salt and IL.

electrolytes are conductive salts.[79] Nevertheless, there are some inconveniences in using them such as low conductivity and specific capacitance, they are also volatile, toxic, flammable, and costly.[80] Hence, they require purification procedures and complex assembly under main environments. Hashemi et al. used a novel regenerative electrocatalytic approach for electrode materials to develop efficient SCs.[81]

2.3.3. Ionic Liquid Electrolytes

Ionic liquid electrolytes are used in supercapacitors to stabilize the electrode materials at high temperatures.[82] On the market, SCs based on organic electrolytes are not appropriate at high temperatures (above 70 °C) due to their low kindling and explosion temperature. In addition, aqueous electrolytes cannot be utilized above 80° and restricted at about 100 °C. Therefore, IL electrolytes are suitable materials used to design high-performance SCs. They have many interesting properties such as electrochemical stability at high temperatures, a wide range of operating voltage, low vapor pressure, non-volatility, and non-flammability. One particularity of ionic liquids is their self-dissociation into cations and anions without any solvents. The ionic liquids are largely used in technology, especially in the hypothesis of green chemistry. In 1998, Anastas and Warner formulated twelve principles of green chemistry to describe the methods of implementation. Ionic liquids follow at least three principles; they are used as (1) safer solvents, (2) they provide energy efficiency and (3) they are used in catalytic reactions. Ionic Liquids (ILs) are chemical compounds based on ion structure, which have a melting point below 100 °C. However, molten salts are characterized by symmetric anions and cations that reinforce the structure and, hence need a high amount of energy to destroy the structure. In contrast, IL cations compared to molten salts, have an irregular shape preventing crystal packing.[82] T Tonic liquids (ILs) are chemical compounds based on ion structure, which have a melting point below 100 °C. The IL structure plays an important role in their properties, which may change by combining cation and anion types (10^{18} possible combinations.[83] However, the change in IL properties is mainly due to the size gap between the organic cation with an extended asymmetric structure and a small organic or inorganic anion. Therefore, this leads to maintaining the disordered state of the atoms in the crystal lattice so consider cutting down the solidification point of the compound. A general model of an irregular system of ions in IL structure is illustrated below in **Figure 9**.

Room-temperature ionic liquids (RTILs) are molten salts at room temperature (below 100 °C), which are also called environment friendly solvents. They are also called environmentally friendly solvents because of their large number of applications in macromolecule chemistry and technology. Seddon and his group have studied the physical properties of ILs to evaluate their purity and the impact of contaminants like water, and chlorine ion.[84]

2.4. Physico-chemical properties of Ionic liquids

ILs are found to be categorized into two types: aprotic ILs (AILs) and protic ILs (PILs).[85] They depend on the nature of the cation and the wise choice of the structure of the ions determines their liquid character. Aprotic ILs (AILs) and protic ILs (PILs) are liquids formed when a proton is transferred between a Bronsted acid and a base. APILs contain no acidic protons.

In contrast, the properties of APILs differ from those of PILs and contain no acidic protons as shown in **Table 2**.

AILs are electrochemically and thermally more stable than PILs and are prominent candidates to substitute volatile and toxic organic compounds (VOCs) due to their ability to be recycled.[86,87] The use of ILs could then solve the problem of solvent loss and traces on the obtained products. The interactions occurring in VOCs are hydrogen bonding, van der Waals, and dipole–dipole interactions. ILs have columbic interactions

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Table 2. Differences between protic ILs and aprotic ILs.

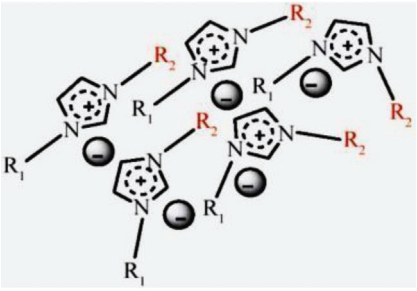
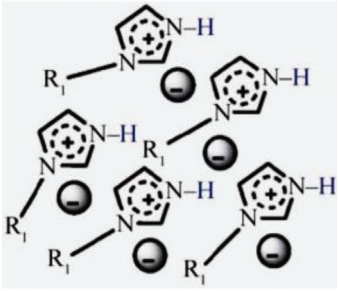
Aprotic ILs	Protic ILs
	
R1, R2 = alkyl group	R1 = alkyl group
low fluidity and conductivity	low melting points
high melting points	high fluidity and conductivity
more expensive and complicated (multistep reactions)	easier to synthesize and low costly (no by-products)
suitable for lithium batteries	suitable for fuel cells

Table 3. Comparison of properties of ionic liquids with those of organic solvents.[88]

Property	Ionic Liquids	Organic Solvents
Number of solvents	>1000 000	>1000
Applicability	Multifunction	Single function
Catalytic ability	Common and tuneable	Rare
Chirality	Common and tuneable	Rare
Vapor pressure	Negligible under normal conditions	Obeys the Clausius-Clapeyron Equation
Flammability	Usually non-flammable	Usually flammable
Solvation	Strongly solvating	Weakly solvating
Tuneability	Unlimited range means designer solvents	Limited range of solvents Available
Polarity	Polarity concept questionable	Conventional polarity concepts Apply
Cost	2 to 100 times the cost of organic solvents	Normally inexpensive
Recyclability	Economic imperative	Green imperative
Viscosity [cP]	22–40 000	0.2–100
Density [g cm^{-3}]	0.8–3.3	0.6–1.7
Refractive index	1.5–2.2	1.3–1.6

(attraction or repulsion) between the opposite charges which allow them to be miscible with polar substances. **Table 3** shows the difference between ionic liquids (ILs) and organic solvents.[88]

ILs can be formed from several hundreds of resulting combinations because of the large number of structures of cations anions. IL cations are generally organic structures that often contain phosphorus or nitrogen atoms with linear chains of alkyl groups such as phosphonium, ammonium, pyridinium, imidazolium, alkylammonium, alkylphosphonium, pyrrolidinium, guanidinium, and alkylpyrrolidinium. IL anions can be from organic anions such as NO_3-, TFSI-, and OTf- or topically from a wide number of inorganic anions which include polyatomic inorganic (PF_6-, BF-), polyoxometallates and halides (Cl^-, Br^-, I^-). The traditional cations and anions used in ILs are shown in **Figure 10**.

The parameters such as viscosity, solubility, hydrophobicity, melting point, conductivity, and so on, can be modified in ILs b replacing the R group with the combined anion or the cation. Coulombic interactions are crucial factors defining the interactions in ILs, which increase the cohesive energy. In reality, all the ionic species which appear to be available do not contribute to the conduction process. Some of them exhibit much lower molar conductivity due to their difference in viscosity (η = cte). Therefore, the ionic conductivity relies strongly on cation-anion interactions and ion sizes. IL is extensively applied in many applications as electrolyte material such as in energy storage devices.

2.4.1. Conductivity

Ionic liquids have moderate specific conductivities. **Table 4** shows various liquids at room temperature.

Ionic liquids (ILs) have conductivities lower than aqueous solutions (KCl and H_2SO_4) and higher than water. They are potential candidates for common electrolytes. The ionic conductivity in

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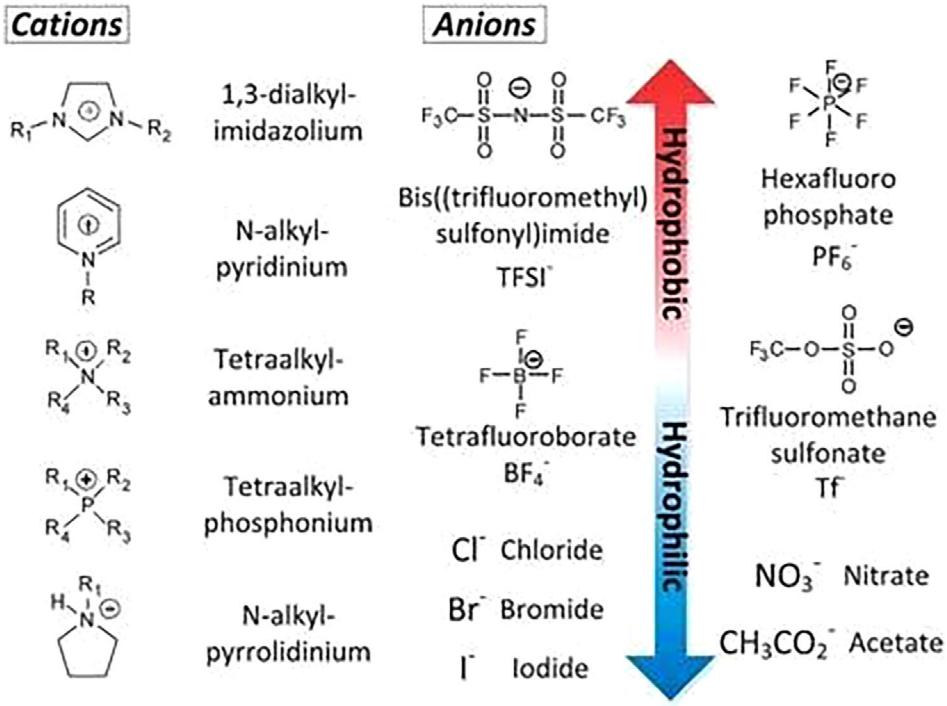

Figure 10. Some common useful cations and anions with their ILs, with their anion-type associated properties. R groups may take on hydrogen atoms or different alkyl groups such as methyl, ethyl, propyl, butyl, and so on.

Table 4. Specific conductivities of liquids at ambient temperature.

Liquids	Specific Conductivity (S/cm)
NaCl	3.88 (T = 900 °C)
KCl (0.1 M aq.)	0.013
H_2SO_4	0.0104
[C2mim][NTf2]	0.0019
[C4py][BF4]	0.0019
H_2O	4×10^{-8}

ILs varies with the diffusion rate of electrically charged particles (ions) through the liquid, which is also high but low if the ions are moving tardy.

2.4.2. Viscosity

This parameter defines the ionic conductivity of ILs. Low viscous ILs exhibit high ionic conductivity, whereas high viscous ILs exhibit low ionic conductivity.

Anions can combine with cations to form weak hydrogen bonds, which may change the IL viscosity. Low viscous ILs exhibit high ionic conductivity because the ions are free to move in the liquid compared to those with high viscosity which exhibit low ionic conductivity. In terms of applications, low viscous ILs are desired for solvents to raise the mass transfer rate whereas higher viscous ILs may be more profitable in the separation process of the sustained membrane/lubrication. Generally, the IL viscosity varies largely with various types of cations and anions. High viscous ILs are those that contain the longest alkyl chain as follows in Equation 3:

$$[CH_3COO_4]^- > [PF6]^- > [C1SO_4]^- > [C2SO_4]^- > [BF4]^- > [OTf]^- > [NTf2]^- \quad (3)$$

It should be noted that the ability effect of anions to combine with cations to form weak hydrogen bonds may change the IL viscosity.[89] [NTf2] –based ILs exhibit low viscosity because they combine better charge delocalization with greater flexibility.[90] The viscosity of ILs is generally related to the symmetric or spherical properties of anions.

2.5. Density

In general, the density goes up with pressure rising and decreases with temperature increasing. Ionic liquids have a density that increases with pressure rising and decreases with temperature increasing. Ionic liquids based on the same cation have a density following this order Equation 4:

$$[BF4]^- < [OTf]^- < [PF6]^- < [NTf2]^- \quad (4)$$

As an experimental result, the density of $[C4mim]^+$ based ILs and various anions such as $[NTf2]^-$, $[PF6]^-$, $[OTf]^-$ and $[BF4]^-$ anions, showed that as the temperature increases the density follows the order Equation 5:[91]

$$[C4mim][BF4] < [C4mim][OTf] < [C4mim][PF6] < [C4mim][NTf2] \quad (5)$$

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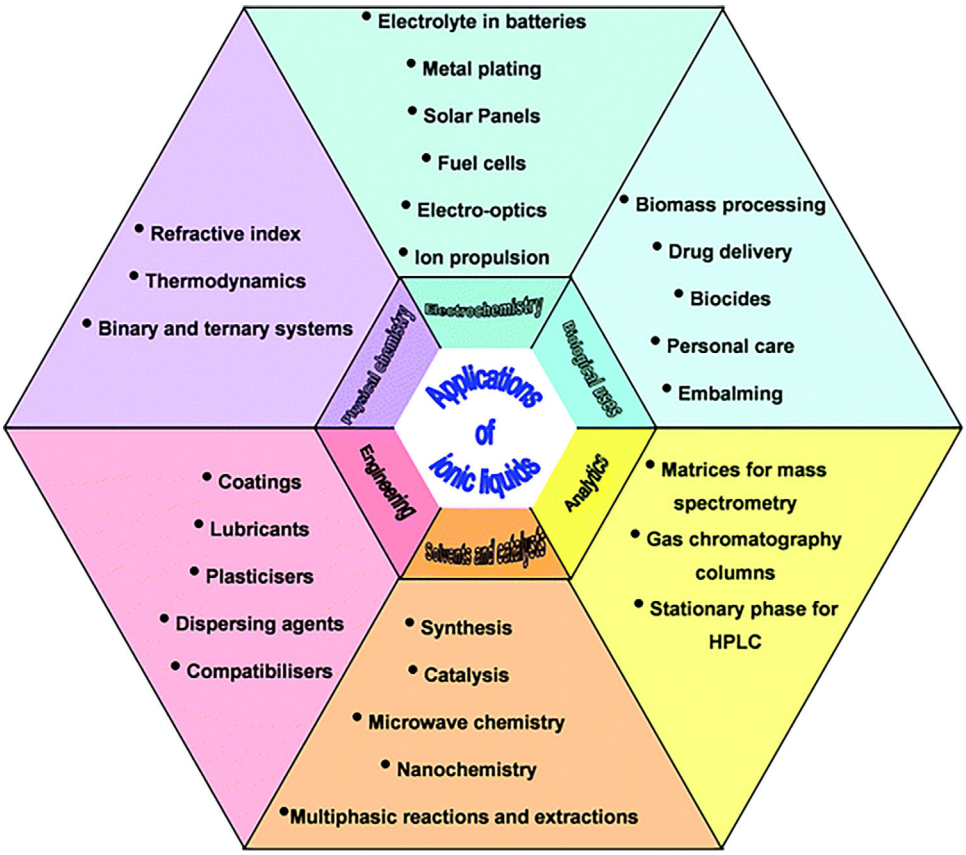

Figure 11. Some applications of ionic liquids.

The density of ionic liquids based on $[NTf2]^-$ as a function of temperature with different cations such $[C2mim]^+$, $[C4mim]^+$,$[C6mim]^+$, $[C8mim]^+$, $[C10mim]^+$, $[CNmpyrro]^+$,$[CNpy]^+$ and $[P6661]^+$ decreases as the temperature increases and following the order Equation 6:[91]

$$[P66614][NTf2] < [C10mim][NTf2] < [C8mim][NTf2] < [C6mim][NTf2] < [C4mim][NTf2][C2mim][NTf2] < [CNmpyrro][NTf2] < [CNpy][NTf2] \quad (6)$$

The physical properties of ILs include those of pure components such as density, viscosity, and melting point, and also blend properties such as phase behavior. A rapid and precise approach to predicting the density of ILs at various temperatures is significant. The approach of molecular dynamics simulation has been developed to predict the liquid density of ILs.[92,93] The phase behavior is a crucial parameter required to understand ILs. **Figure 11** shows the application of ionic liquids.

ILs compared to conventional solvents, are non-volatile, low toxic compounds, easy to manipulate, non-flammable, and with high ionic conductivity. For all these reasons, they are widely used as green materials in polymer chemistry and technology.

2.6. Advantages of Ionic Liquids

2.6.1. Ionic Liquids as Moderators

MacFarlane and his co-workers used ILs as moderators in the exothermic polymerization of styrene and acrylonitrile.[94] This led to diminishing decomposition and exothermic self-heating of the product. IL reacts with methanol in a closed cycle and accelerates the polymer recovered by IL via the evaporation of the methanol solution.[95]

2.6.2. Ionic Liquids as Stabilizers

Mecerreyes et al. used PILs such as (Pm [VEIM][Br]) in the phase transfer, as steric stabilizers to develop a new method for the preparation of intrinsic conducting polymers (ICPs) organic dispersions.[96] PILs can be used as smart stabilizers for metal nanoparticles as shown in **Figure 12**.

Ionic liquids (ILs) have been used as smart stabilizers for metal nanoparticles in the synthesis of polypyrrole (PPy) dispersions.[97] Water-soluble ILs (WSILs) were used by Schubert et al. as stabilizing compounds with their surface-active- properties without polymerization reactions.[98]

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Figure 12. Poly (ionic liquids) as "smart" stabilizers for metal nanoparticles.

2.6.3. Ionic Liquids as Modifiers

Jesionowski et al. used ILs to augment the density of bulk silica and Ding et al. used ILs to modify the properties of polypropylene melt intercalation.[99] The density of bulk silica was augmented using IL on its surface. The use of ILs in the silica change showed great benefit of effective surface hydrophobization. Ding et al. used IL ([C18MIM][Cl]) as a modifier via ion exchange from sodium montmorillonite in toluene into the synthesis of a series of organ- montmorillonites (O-MMT) and studied its influence on polypropylene (PP) melt intercalation.[100] They found that MMT layers absorbed [C18MIM] cation through mimetic physical and chemical adsorption; then contributed to changing some of [C18MIM] cations into chemical adsorption as the loading level of ([C18MIM][Cl]) increased.

2.6.4. Ionic Liquids as Lubricants

Ionic Liquids (ILs) are new promising lubricant additives with great beneficial potential such as enhanced wear protection, no device efficiency loss, improved engine durability, and extended service intervals, leading to better fuel economy using less viscous oils.[101] RTILs were employed by Bermudez et al. as internal and external lubricants into novel polymer-IL dispersions and poly (styrene)/PA6-steel interfaces respectively.[102] They exhibit low friction and are hugely soft in poly (styrene)/AISI 316L rustproof steel contacts which were developed regardless of IL composition. There were no tribocorrosion processes when using ISI 316L stainless steel pins but appeared with the use of ILs as external lubricants of PSt/AISI 52 100 contacts. The glass transition temperature (Tg) of poly (styrene) was increased by IL dispersed phases while softening PA6 and diminishing its temperature of decomposition. The short alkyl chain ([EMIm][BF4]) was the first IL to be employed as an inner lubricant in epoxy resin in a range of concentrations [0.5–3.3 wt%].[103]

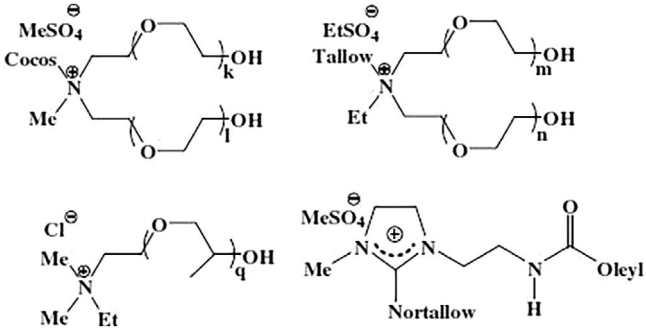

Figure 13. Commercially available unconventional ILs.

2.6.5. Ionic Liquids as Performance Additives

ILs were utilized in various materials as performance additives and manners were reported by Weyershausen and Lehmann.[104] Ionic Liquids (ILs) were utilized in various materials as performance additives, and unconventional ILs provided an interesting and attractive solution to the industry of painting and coating. In addition, the use of ILs during storage and manufacture reduces the complexity of logistics. **Figure 13** shows unconventional ILs used as performance additives for industrial applications.

2.6.6. Ionic Liquids as Surfactants

Yan and Texter revealed that using surfactant ILs, polymer particles, and open-cell microporous, gel materials can be produced from Bulk microemulsion polymerizations.[105] In this process, polymer particles, open-cell microporous, and gel materials can be produced from Bulk microemulsion polymerizations. They found that the change from the hydrophilic nature to the hydrophobic nature in copolymer at aqueous/polymer domains interfaces was due to the anion exchange from [Br] to [PF6] and making pores at the interface by shrinkage.

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2.6.7. Ionic Liquids as Catalytic Agents

Ionic liquids have low viscosity, good conductivity, excellent thermal stability, and good stability potential window, and are actual novel materials for developing novel highly conducting electrolytes for lithium-ion batteries and SCs. Nevertheless, ILs present some drawbacks in the practice such as their fluidity which may cause liquid leakage. The polymerization of ILs composed of imidazolium-substituted methacrylates was attempted by Ohno and co-workers.[106] However, polymerizing ionic compounds reduces the motion of the polymer chain and then reduces its conductivity. The solution could be found in three-dimensional highly crossed-linked polymers with excellent physicochemical properties. It was reported that the incorporation of 50 wt% of IL([DMIm][Br]) into the epoxy matrix formed highly thermally stable and solid electrolyte with good conductivity of around 10^{-3} S m^{-1} at 25 °C and 0.1 S m^{-1} at 170 °C.[107] Solid electrolytes have good potential to be used at elevated temperatures in energy storage devices. Its good flexibility solved the leakage problems because of its low glass transition temperature.

2.6.8. Ionic Liquids as Plasticizers

Plasticizer material is the one used in a polymer to enhance its properties such as flexibility, functionality, or distensibility. They modify many properties of polymers such as the reduction of their melt viscosity, and glass transition temperature (T_g). In the industry, the most used plasticizers are made from phthalates (70% of the commercial market). However, the major problem of these plasticizers is their toxicity. For this reason, many researchers have been searching for novel plasticizers. They found that RTILs have excellent properties; they are non-volatile at ambient conditions. The organic cation and inorganic anion of RTILs both have an important effect on the distribution of ionic regions in the polymer matrix. RTILs are non-volatile at ambient conditions, and they have excellent properties such as decreasing the melt viscosity, and glass transition temperature, and improving the mechanical properties of thermosets.[108] Lu et al.[109] reported that using several types of ILs, T_g of epoxy networks decreased significantly. This affected the modified systems with imidazolium-based ILs rather than systems based on phosphonium. Sanes and coworkers[103] showed that epoxy resins doped with IL increase the chain motion of polymer and softening as well as reduce the wear rate and friction coefficient of the softened materials. Brazel and his co-workers used [BMIM][PF6] and [HMIM][PF6] as plasticizers for PMMA.[110] These plasticizers reduced T_g to nearly 0 °C and also improved thermal stability than dioctyl phthalate (DOP) plasticizers. They can be used to replace flexible polymers at high temperatures without losing mechanical strength or even brittleness. Rahman and Brazel found that RTILs based on imidazolium-, phosphonium- and ammonium cations as plasticizers can produce flexible PVC as does DEHP.[111] They revealed that RTILs based on imidazolium, phosphonium, and ammonium cations as plasticizers incorporated into PVC formed good flexible plastic, and many ILs doped PVC show better flexibility compared to traditional plasticizers. The behavior of IL as a plasticizer exhibited better migration resistance and leaching than traditional plasticizers when they were exposed to far-range UV.

2.7. Ionic Liquid for Supercapacitor Devices

Electrochemical devices such as supercapacitors can aid in reducing the use of fossil fuels in everyday life. Mysyk et al.[112] claimed that the energy density is higher in SCs than electrolytic capacitors and the power density in SCs is higher in SCs than rechargeable batteries. The three general classes of supercapacitors: are capacitors (EDLCs), pseudocapacitors (PCs), and hybrid capacitors and the difference between them is the charge storage mechanism. In practice, pseudo-capacitance, usually involving proton and electron exchange between the electrolyte and the electrode, is exploited in systems based on low-cost aqueous electrolytes[113] Supercapacitors consist of components such as a cathode, electrolyte, and cathode. The working potential of a supercapacitor highly relies on the ESPW of the electrolyte. In a supercapacitor, the electrolyte and electrode also establish its cyclability and power density. Hence, a good electrolyte for supercapacitor application is required to enhance the performance. Researchers have been attracted by carbon-based capacitors because the carbonaceous materials exhibit excellent cycling stability, various morphologies, high power capability, and high conductivity.[114] The ionic liquid DEME-BF4 shows considerable conductivity of 4.8×10^{-3} S cm^{-1} at 25 °C and also with a high potential window of 6 V in electrochemical capacitors.[115] Yuyama and his coworkers.[115] reported that the stability and performance of $BF4^-$ anion-based IL used in EDLCs were better compared to those that are composed of $PF6^-$ and $TFSI^-$ conductivity. This result was assumed by IL which plasticized the system in reducing the Tg. The plasticization effect of IL enhances the amorphousness of the polymer which increases the flexibility of the polymer backbone. This aids the ions transport and enhances the ionic conductivity. In addition, an IL-doped polymer electrolyte containing an ion-conducting medium increased the amorphousness of the polymer membrane and exhibited a wider range of ESPW. The electrochemical characteristics of the EDLC were 2.39 W h kg^{-1} and 19.79 W kg^{-1} for specific energy and power respectively. The addition of RTIL to a polymer matrix is another approach to get good electrochemical and thermal stability.[116] Pandey and coworkers[117] have investigated the performance of both solid-state electrolyte-based EDLC and IL-doped polymer electrolytes. They fabricated the EDLC electrodes with MWCNT and the polymer electrolyte was a combination of PEO, $(CF_3SO_3)_2$ Mg, $LiCF_3SO_3$, and IL (EMImTf). The room temperature ionic conductivity was about 10^{-2} S m^{-1}. By doping IL into the polymer electrolyte (PE), the flexibility of the polymer backbone chain increased and thus, enhanced its segmental motion. The presence of various free ions such as Li^+/ $Mg2^+$, $EMIm^+$, and $CF3SO3^-$ in PE multiplied the chance of obtaining diverse double layers at the electrode-electrolyte interface. EDLC could deliver a specific capacitance of 1.7-2.1 F g^{-1} using MWCNT/Li-based and 2.6-3.0 F g^{-1} using MWCNT/Mg-based electrolyte. In conclusion, the incorporation of IL in the EDLC has enhanced its capacitance. The incorporation of BmImCl into the PVA: CH COONH anions. They demonstrated that $TFSI^- > PF6^- > BF4^-$ as the ESR and direct current (dc) resistance decreases at 298 °K and 243 °K.

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Fletcher and his coworkers[118] claimed that the decrease in IL electrolyte viscosity with increasing temperature suggests the decrease of direct current (dc) resistance and ESR. This led to an increase in cation and anion diffusion and this implies an increase in ion dissociation and mobility at elevated temperatures. Like AILs, PILs show a high stability potential window when utilized with non-porous materials.[119] This type of ionic liquid compared to AILs is inexpensive and easy to prepare.[120] PIL-based capacitors exhibit a wider ESPW than aqueous H_2SO_4-based capacitors. The combination of PIL and AC is a promising candidate with good energy characteristics for supercapacitor applications. Brandt et al.[121] investigated several supercapacitors based on carbon with different PILs such as Me3NHTFSI, PYRNO3, and Et3NHTFSI as electrolytes to enhance their cycling stability. As a result, they reported that PILs-based supercapacitors exhibited high stability (30000 cycles) and they can also operate at high temperatures. Many researchers have investigated the performance of IL electrolytes combined with different cations such as BMP^+, $EMIm^+$, and anions such as $BF4^-$, $TFSI^-$, and DCA^-.[122] Amongst all these combinations, IL ([BMP][DCA]) exhibits good electrical behavior of the electrolyte using graphene nanosheet (GNS) based electrodes for supercapacitor application. The addition of 50 wt% [BmIm][Cl] to the PVA/CH_3COONH_4 electrolyte system[123] at 393 °K leads to achieving 0.73 S m^{-1} of polymer salt complex improved the conductivity, because of its large numbers of ions, excellent properties of plasticizer, and environmentally friendly nature.[124] The presence of IL in the polymer electrolyte contributed to the flexibility of the polymer chain and thus increased the motion of the polymer segment. This is conducted to higher the ion transfer and ionic conductivity. The differential scanning calorimetric (DSC) analysis confirmed this result by observing the addition of IL.

Moreover, IL has physicochemical properties such as dielectric constant and viscosity which strongly participate in enhancing ionic conductivity. IL has physicochemical properties that enhance ionic conductivity and enhance PE and EDLC properties. It is a crucial method to ameliorate the performance of EDLCs. Therefore, the inclusion of IL into the PE is a crucial method to ameliorate the performance of EDLCs.

2.7.1. Synthesis Method to Develop IL-based Polymer Electrolytes

In everyday life, polymers are a very important and useful class of materials in science and technology. They can be available in diverse forms such as in rubber, resins, plastic, adhesives, and adhesive tapes. Polymer is composed of two Greek words "POLY" and "MEROS" meaning "many" and "parts" respectively. Polymers are very large molecules made up of smaller units (monomers), covalently bonded together. The combination of monomers together by the reaction of polymerization forms polymers.[125] Human beings used natural polymers in the fabrication and design of clothes, tools, writing materials, weapons, shelter, and other important materials. In the eighteenth century, the scientist called 'Thomas Hancock suggested a way to have a natural rubber modified through blending with certain additives materials and Charles Goodyear proposed the process of vulcanization to get the properties of natural rubber enhanced with sulfur. Premium polymers such as synthetic polymer and synthetic fiber were produced in 1909 and 1911 respectively. The science of polymer started in the twentieth century with Herman Staudinger who was the first to publish the concept of high molecular mass compounds. Ionic liquid-doped polymer electrolytes are promising candidates to achieve higher ionic conductivities and can replace other solvent-free systems. Thermally stable ILs are widely used because of their unique properties such as non-flammability, non-volatility, good potential proton conductivity, high ion density, and excellent ionic conductivity.[126] These properties play a crucial role in gel or solid electrolyte matrixes in electrochemical devices. On the other hand, ILs are suffering from fluidity. Extensive research on the IL-doped polymer electrolytes reported in the literature has revealed that the transport properties of polymer electrolytes can be crucially improved by the incorporation of ILs upon controlling the degree of ion dissociation, the concentration of ionic moieties, and Tg of the membranes.[127] The concept of 'salt-polymer was developed by Angel et al., to solve this problem to achieve highly conducting polymer electrolytes.[128] The development of IL-incorporated polymer electrolytes can be designed using the combination of ILs and conventional polymer matrices or to design functional polymers promoting some of the IL characteristics.

2.8. IL Doping into Polymer

2.8.1. Impregnation Synthesis

In this approach, an IL-doped polymer electrolyte is obtained by impregnation of the polymer membrane into an IL solution. The polymer electrolyte can be also obtained by solution casting technique using various kinds of polymers such as homopolymer, copolymer, block polymers, or polymer blend solutions to obtain porous or dense membranes[129] and utilizing non-covalent or covalent cross-linking methods.[130] This method shows some drawbacks such as the limited choice of polymers and ILs and its inability to modify interactions between the IL phases and the polymer. Although a limited amount of IL is present in the polymer membrane, it has been observed that the selection of appropriate ILs and polymer matrices or augmenting the temperature can attain high proton conductivity. Doyle and his team have prepared Nafion-based IL membranes that offered high conductivity up to 110 mS cm^{-1} at 180 °C [IL uptake up to 120% (~54 wt % IL content)].[131] Different methods of preparation are illustrated in Figure 14.

2.8.2. Solvent Blending Synthesis

This approach consists of preparing a polymer electrolyte by mixing IL homogeneously with a synthesized polymer in a chosen solvent. The homogeneous solution is poured into a mold and then dried to remove all the solvent molecules for a good solidification of IL. This method offers a quick way to control the conductivity by varying the volume of IL and the type of polymeric materials in composite membranes.[132] The content of IL is therefore an important factor that will modify the properties of the obtained IL-doped polymer gels such as its conductivity. However, an excess amount of IL diminish mechanical stability and IL leaching.

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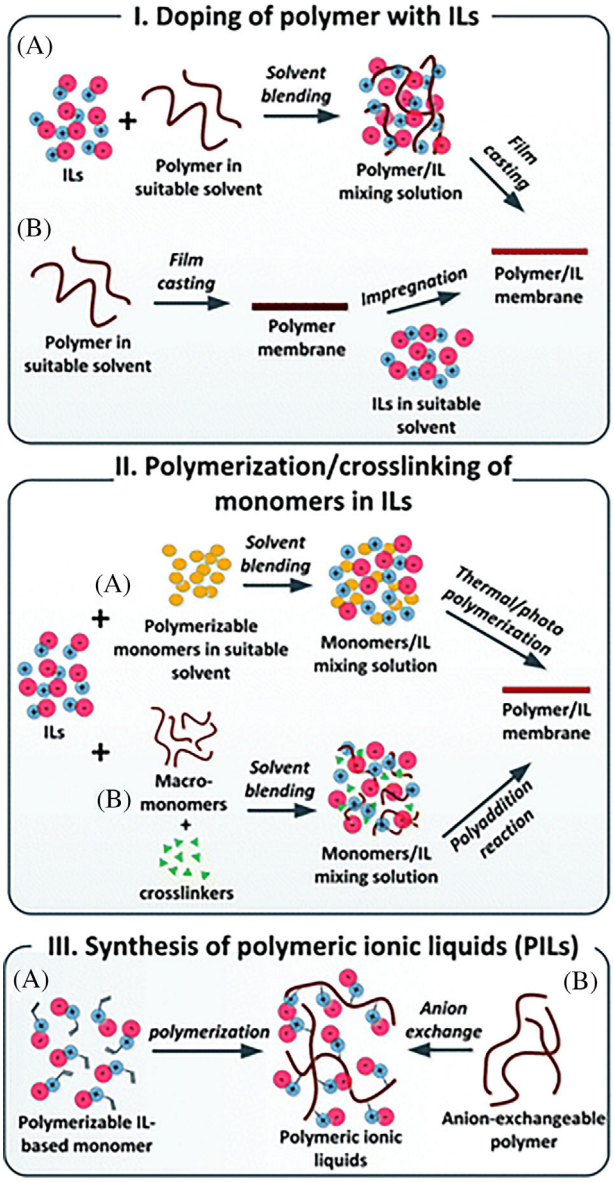

Figure 14. Preparation of IL-doped polymer electrolytes.

This generally happens when the polymer is poorly compatible with the IL. Therefore, the good preparation of ILDPEs depends on the compatibility between IL and polymer matrix ratio.[132]

2.9. Modification of the Conductivity in Polymer Electrolyte by IL

2.9.1. PEO + $NaNO_3$: IL System

Sandhya Gupta and her co-workers[133] prepared and characterized different electrolyte systems using PEO, NaNO3, and [EMIM][SCN]. They performed DSC and IS to check the percentage of crystallinity and electrical properties, respectively. Doping IL into polymer increased its conductivity. The maximum conductivity reached 1.93×10^{-4} S cm^{-1} with 9wt%IL. DSC thermograms revealed a change in IL-doped PEO: $NaNO_3$ (95: 05) film. They found electrical behavior of polymer electrolytes without IL strongly depends on the number of charge carriers whereas mobility controls that of polymers with IL. The number of charge carriers and mobility play an essential role in the conductivity of polymer without and with IL respectively.[134]

2.9.2. PVDF-HFP: [EMIM][TCM] Polymer Electrolyte System

Tuhania et al.[135] used ILDPE in EDLC fabrication and achieved a high conductivity and specific capacitance utilizing the optimized polymer electrolyte and corn starch-based porous carbon electrodes. They used P (VDF-HFP) and IL ([EMIM][TCM]) as a dopant. EIS have been performed and the highest conductivity was 2.65×10^{-3} S cm^{-1} for 80 wt% IL-doped electrolyte film. LSV has been performed using the optimized electrolyte and two stainless steels. The stability potential window was 3 .6V. POM analysis of the prepared films shows a reduction in crystallinity due to the IL. IR spectrum further confirms its composition nature. The electrical behavior of dielectric response and conductivity is in the same trend. The specific capacitance *Csp* of the EDLC was 2.36 F g^{-1} utilizing the optimized polymer electrolyte and corn starch-based porous carbon electrodes.

2.9.3. Ionic Liquid-Based Gel Polymer Electrolyte

Lee et al.[136] have prepared novel IPGEs using ionic liquid (Mor1,2 TFSI) and P(VDF-HFP) copolymer. The conductivity of IPGE films was studied with and without propylene carbonates (PC) with increasing temperatures at different amounts of [Mor1,2][TFSI]. As a result, the conductivity rises as the temperature and volume of IL increase. The conductivity of IPGEs increases with increasing temperature and volume of IL, but they are thermally unstable. The maximum conductivity of IPGE film was found to be 1.1×10^{-2} S cm^{-1} at 60 °C, but they are thermally unstable.

2.9.4. Experimental Investigations of an IL-Based Mg^{2+} Conducting Polymer Gel Electrolyte

Hashmi et al.[137] prepared a novel magnesium-doped gel polymer electrolyte based on P(VDF-HFP) and IL (EMITf) that demonstrated good properties as electrolytes in rechargeable magnesium batteries. The electrolyte system consists of Mg-based salt [Mg $(CF_3SO_3)_2$] incorporated into P(VdF-HFP) which is semitransparent and freestanding with excellent mechanical strength and film flexibility. They studied the electrochemical and physical properties of these films which demonstrated good properties as electrolytes in rechargeable magnesium batteries. The electrolyte exhibited good electrochemical and thermal stabilities with conductivity of about 4.8×10^{-3} S cm^{-1} at 20 °C. FTIR showed a change in the polymer electrolyte structure by IL doping. XRD and SEM techniques confirm a decrease in crystalline nature. The different approaches CV, EIS, and TNM confirm the transport of Mg^{2+} in the polymer gel film. The ionic transference number (t^+) was found to be 0.26 which demonstrates an important input of triflate anion in the conductivity owing to the presence of IL.

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2.9.5. Incorporation of EMITf into Mg/Li–PEO for EDLC Supercapacitor

The addition of IL to Mg/Li – PEO as a polymer electrolyte enhances the EDLC performance in both cases (Mg/Li) and their ionic conductivity increases as temperature increases. Hashmi and al. used MWCNT-based electrodes to fabricate Mg and Li-based EDLC.[138] They found the maximum conductivity of polymer electrolytes to be 10^{-4} S/cm at ambient temperature and 10^{-3} S/cm at about 100 °C. The Mg- ion-based polymer electrolyte shows relatively better performance than the Li-ion-based system. The Li^{+}-based polymer electrolyte delivered a stability potential window of ~4 V and slightly higher for the based $Mg2^{+}$ polymer electrolyte. The Mg-based EDLC shows a specific capacitance value of 2.6–3.0 F g^{-1} and 1.7–2.1 F g^{-1} for Li-based cells.

2.9.6. PVA + CH_3COONH_4/BmImTf System

Liew and coworkers[139] used the solution casting technique to prepare ion- ion-ion-conducting polymer electrolytes using polymer PVA/ammonium acetate (CH_3COONH_4). The addition of the IL (BmImI) enhanced the thermal stability of the polymer electrolyte and also its stability potential window up to 3.8V. The EDLC cell was built with the maximum conducting polymer electrolyte film. The specific capacitance was higher than that of the EDLC based on IL-free polymer electrolyte. This demonstrates IL in polymer enhances the performance of the capacitor device. The electrochemical parameters such as specific capacitance, power, and energy decrease as the cycle number increases. This polymer electrolyte is a sustainable material to be utilized in EDLC devices to ameliorate their electrochemical performance with excellent coulombic efficiency. PVA has the potential to be applied in electrochemical energy conversion devices, such as batteries, alkaline fuel cells, and electrolyzers.[140]

3. Conclusion

In this research work, we studied the components and materials used in the supercapacitor and the synthesis process of solid electrolytes for EDLC applications. Ionic liquids are promising candidates to enhance the electrochemical performance in EDLC supercapacitors. Ionic liquids are promising candidates to enhance the electrochemical performance in EDLC supercapacitors. IL acts as a source of ions to augment the number of ionic charges and improve ion transport in polymer electrolytes. The amorphousness of 19 SPE can be improved by using IL which leads to conductivity enhancement by increasing the flexibility of the polymer chain. Low viscosity IL contributes to improving the ion transport, the mechanical and thermal stability. IL can be used as a separator between the two electrodes or like the electrode-electrolyte interface contact which contributes to high energy storage and capacitance of electrode materials.

Ionic liquids are not simple fluids, because they can have multiple targeted combinations that modify almost physicochemical properties of electrolytes. ILs possess some advantages such as offering a large number of applications in science and technology due to their high electrical conductivity, high thermal stability, wide potential windows, low vapor pressure, and non-flammable. Ionic liquid-doped polymer electrolytes show high conductivity and other useful properties as expected by a good polymer electrolyte. We have reported the performance of some polymer electrolyte systems prepared for electrochemical devices such as EDLCs. This chapter also talks about the history, the mechanism of energy storage of EDLC, and the importance of low-viscosity ionic liquid in EDLC supercapacitors. The use of a separator membrane is not necessary between the two electrodes, leads to cheapening the price of EDLC cells.[141–144]

Acknowledgements

The corresponding author is thankful for the support and cooperation from their host institutions, and his co-author (Sandeep Kumar) is thankful to the CSTUP (CST/D-1041) for their financial assistance.

Conflict of Interest

The authors declare no conflicts of interest.

Author Contributions

Conceptualization: Azemtsop Manfo Theodore. Software: Azemtsop Manfo Theodore. Validation: Azemtsop Manfo Theodore. Writing—review and editing: Azemtsop Manfo Theodore.

Data Availability Statement

The data that support the findings of this study are available from corresponding author upon reasonable request.

Keywords

capacitance, electrode material, electrolyte, ionic liquid, polymer electrolyte, supercapacitor

Received: February 28, 2024
Revised: April 3, 2024

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